Time-resolved x-ray diffraction from laser-irradiated crystals

J. Larsson,[1] P.A. Heimann,[2] A. Lindenberg,[1] P.J. Schuck,[1] P.H. Bucksbaum,[3] R.W. Lee[4], H. A. Padmore,[2] J.S. Wark,[5] and R.W. Falcone[1,6]

[1]Physics Department, University of California at Berkeley, Berkeley, CA 94720

[2]Advanced Light Source, Accelerator and Fusion Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

[3]Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, MI 48109

[4]Lawrence Livermore National Laboratory, Livermore, CA 94551

[5]Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, U.K.

[6]Center for Beam Physics, Accelerator and Fusion Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

INTRODUCTION

The properties of semiconductors following irradiation with ultrashort pulse lasers has been studied extensively with optical probes.[1-2] One motivation for this work has been the determination of the mechanism of ultrafast disordering (melting). By probing laser-irradiated material using ultrafast time-resolved x-ray diffraction, it should be possible to directly determine the mechanism for the transition from order to disorder on ultrafast timescales.

In our experiment we studied the disordering of laser-illuminated InSb. The experimental setup utilizes ALS beamline 10.3.2 and is described in more detail in Ref. 3. X-rays are monochromatized and focused to a line using a bent silicon wafer cut in the (111) plane. The Bragg angle is chosen to be 22.5 degrees which gives a photon energy of 4.8 keV for Si (111). A Ti:Al_20_3-based laser system produced pulses with a duration of about 100 fs at a repetition rate of 1 kHz; it has been synchronized to the electron storage ring with jitter less than 10 ps. The laser beam is focused on a InSb (111) sample at a fluence of about 30 mJ/cm^2, about a factor of two below the threshold for rapid and visible damage. Diffracted x-rays are detected by a x-ray sensitive avalanche photodiode (APD).

In InSb photoabsorption limits the x-ray penetration depth. We note that the linear absorption coefficient at the 800 nm laser wavelength corresponds to a attenuation length of about 100 nm. The x-ray probe depth (at 5 KeV) is about 200 nm.

RESULTS AND DISCUSSION

Using a pump-probe technique, the diffracted x-ray intensity is observed as a function of scanned delay between the laser heating pulse and the x-ray probe. A 30 % drop in the diffraction intensity is seen with a time-resolution limited by the ALS-pulse duration, which is on the order of 70 ps for the operating conditions used. Then, the diffraction efficiency recovers over a 100 ns timescale.

In Fig. 1, we show theoretical and experimental rocking curves with different delays between the laser heating and x-ray probe. The calculation numerically solves the dynamical diffraction equations. We set the initial temperature distribution to be an exponential with a decay depth equal to the laser absorption depth, where the top diffracting layers are at the melting temperature. In comparing experiment and theory we can explain the shape of the rocking curve quite well.

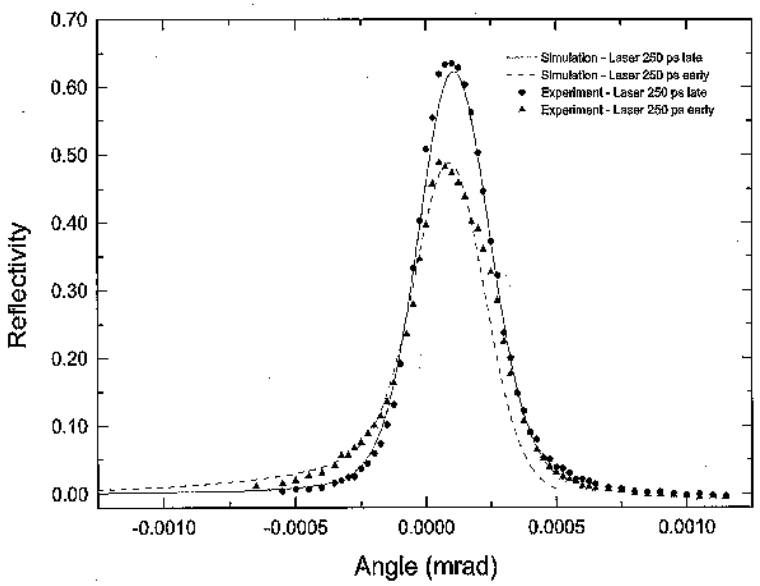

Figure 1. Experimental and theoretical rocking curves for the (111) reflection of InSb.

A change in the diffraction efficiency can be related to several possible effects including: lattice expansion, melting with regrowth, or a phase transition to a different structure. We interpret the data as follows. As the laser interacts with the surface, the top layer (40 nm thick) melts causing a drop in the diffracted intensity. The underlying layers are heated resulting in a strained lattice with increased crystal spacing. Then, the lattice regrows.

In order to access a time resolution beyond the ALS pulse duration, a cross-correlation technique was developed requiring two laser-irradiated crystals. A time derivative of the cross-correlation data shows a change in the x-ray diffraction efficiency of InSb on a timescale of < 2 ps. In conclusion, we show evidence of a laser-induced phase transition in InSb through the study of time resolved x-ray scattering from a laser illuminated crystal.

REFERENCES

1. C.V. Shank, R. Yen and C. Hirlimann, Phys. Rev. Lett. **50**, 454 (1983)
2. I. L. Schumay and U. Höfer, Phys. Rev. **B 53**, 15878 (1996)
3. J. Larsson, Z. Chang, E. Judd, R.W. Falcone, P.A. Heimann, H.A. Padmore, P.H. Bucksbaum, M. Murnane, H. Kapteyn, R.W. Lee and J.S. Wark, Optics Letters **22**, 1012 (1997).

This work was supported by an Academic Research Instrumentation grant from the National Science Foundation, and by the Department of Energy through both Lawrence Livermore National Laboratory and the Lawrence Berkeley National Laboratory.

Principal investigator: Roger Falcone, University of California, Berkeley. Email: rwf@physics.berkeley.edu.